Effect of Thermal Alteration on Petrographic
Constituents of the Hanna No. 1 Coal

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INTRODUCTION

The United States Department of Energy, Laramie Energy Technology Center, (LETC) has conducted numerous underground coal gasification (UCG) experiments. DOE recognizes the great potential value of UCG as a means of extracting energy from coal beds which cannot otherwise be utilized. Many beds are too deeply buried to be strip-mined, or are unsuitable for underground mining, yet constitute a vast energy reserve. We are currently developing UCG technology which should lead to an energy- and cost-efficient means of utilizing such coal beds.

LETC has chosen the Hanna No. 1 coal for experimental UCG work. The Hanna No. 1 lies within the Paleocene Hanna Formation and is confined to the Hanna Coal Basin of Wyoming (Figure 1). The No. 1 coal has been classified as a high-volatile C bituminous coal (1).

Various approaches have been used to improve the efficiency of the UCG process at the Hanna site. While these include proper installation and use of surface facilities, LETC personnel have also recognized the need for a complete understanding of the coal itself. Through careful analysis of the coal, improved systems for conversion to high quality gaseous fuel can be designed. Various types of analyses have been performed on samples of altered and unaltered coal and carbonaceous shales from the Hanna site. This report details the results of one aspect of those analyses.

PURPOSE OF THE STUDY

The purpose of the study has been to construct a geothermometer based upon visible thermal alterations in the organic constituents of Hanna No. 1 coal and carbonaceous shale samples. Youngberg, at LETC, and Rich, at the South Dakota School of Mines and Technology (SDSM&T) agreed that a correlation might be made between discrete alteration temperatures and unique petrographic compositions produced by alteration. If such a correlation could be made, then samples of coal and/or carbonaceous shale taken from UCG sites could be observed microscopically and determinations made as to the maximum temperatures to which the sediments had been subjected. The organic constituents of the samples, termed macerals, were observed, described, and quantified as changes occurred in their appearances during heating at progressively higher temperatures.

LABORATORY METHODS

Sample Preparation

Crushed bulk samples of coal and carbonaceous shale from the Hanna 170 sample site were sent to SDSM&T. One sample from 277-278 feet depth was chosen for a low ash sample (12.7% dry basis), while the sample interval 279.7-280.7 feet was

chosen for a high ash sample (63% dry basis). A third sample interval, 284.2-285.2 feet was added to the work schedule about midway through the project. Its unusual petrographic composition indicated it might actually perform better as an indicator of thermal alteration than the original samples.

Five gram samples were placed in a previously weighed crucible, and their combined weight was recorded. The furnace, meanwhile, had been heated to the desired temperature and thoroughly flushed with helium. Helium was chosen because it is totally inert and would not cause alterations in the samples in and of itself. Heating was done at 50°C intervals between room temperature (~20°C) and 600°C.

After being heated for 1 hour, the crucible was removed from the furnace as quickly as possible and weighed to record any weight change. Each sample was mixed immediately with APCO adhesive #5823. This is a low viscosity epoxy-type embedding medium which is commonly used for coal pellet preparation. The mixture of coal or shale and epoxy was divided between two steel 1-inch internal diameter steel molds, and compressed at 7000 psi for 2 minutes. After each sample hardened (~18 hours) it was labeled, then ground and polished on a Jarret Automatic Grind-Polisher.

Furnace Construction

The furnace apparatus consisted of two Parr oxygen bombs connected by a length of teflon-lined steel mesh hose (Figure 2). Only one bomb was used as a furnace. The furnace bomb was wrapped with Samox-insulated heating tape, which was then covered with several windings of Glaspun fiberglass insulating tape. The heating tape was plugged into a variable transformer which provided current for heat generation.

The lid of a Parr oxygen bomb is equipped with several fixtures, including one-way gas valves which allow flow either into or out of the bomb, and two electrical terminals which, under ordinary oxygen bomb operation, provide a heat source for sample ignition. The inlet gas valve on the furnace bomb was connected to a helium tank, while the exit valve was connected to the steel mesh hose. The hose was also connected to the second bomb, as previously mentioned. The stream of helium, then, flowed into the furnace bomb, out through the hose, and into the second bomb. From there, gases were exhausted into a fume hood. The second bomb served as a barrier to atmospheric gases which might have accidentally entered the furnace bomb, and therefore helped to prevent unwanted or dangerous combustion of samples.

One of the two electrical terminals in the furnace bomb lid was removed, and a thermocouple inserted in its place. The thermocouple was wrapped with fiberglass insulation to provide a tight seal against the air outside the bomb. The end of the thermocouple was inserted to a level just above and to the side of the crucible (Figure 2) in order that the heat sensor would measure the furnace temperature at the sample location within the bomb. Accurate temperature monitoring was provided by an Omega digital centigrade thermometer which was attached to the thermocouple.

The crucible was placed within a circular loop of wire which is designed to hold more conventional oxygen bomb samples. The crucible rested in about the middle of the furnace chamber and was, presumably, heated evenly from all sides.

A typical sample run consisted of the following steps:

1. heating of furnace bomb to preselected temperature

2. flushing of both bombs with helium

3. recording of sample weight

4. insertion of crucible into furnace bomb

5. securing furnace bomb lid

6. heating in a helium stream for 1 hour

7. removal of furnace bomb lid and sample

8. weighing of sample

9. immediate mixing of sample with epoxy

10. pellet pressing and polishing

Sample Analysis

Methods of analyzing polished pellets, or briquettes of coal are fairly well established. Stach (2) presents a concise review of techniques. The method of microscope analysis employed in this studied involved incident illumination of polished pellet surfaces and observation at 500 X magnification in oil immersion. Blue-light (actually blue-and ultraviolet wavelengths) was employed initially, though one typically observes samples using incident "white" light. Such "white" light analyses were provided by LETC, however, and so were not duplicated by Rich for unheated samples. The average maceral compositions of 108 unaltered samples of Hanna No. 1 coal are shown in Table 1.

TABLE 1. Average Maceral Composition (mineral-matter free basis) of 108 Unaltered Coal Samples from the Hanna UCG site (1).

MACERAL GROUP	VOLUME %	MACERAL	VOLUME %
Vitrinite	91.8	Vitrinite	90.3
		Pseudovitrinite	1.5
Liptinite (Exinite)	6.1	Exinite	5.5
		Resinite	0.6
Inertinite	2.1	Semi-Fusinite	1.0
		Semi-Macrinite	0.1
		Fusinite	0.3
		Macrinite	0.1
		Micrinite	0.5
		Sclerotinite	0.1

The application of blue-light was done at first merely to see how many of the macerals in the Hanna 170 samples would fluoresce. Incident blue-light causes liptinite macerals (waxes, resins, spores, etc.) to fluoresce brightly, especially in low-rank coals. As the liptinites fluoresce they can be easily identified and quanitifed.

Sample surveys revealed a particular type of liptinite, exudatinite, to be unusually common in the Hanna 170 core samples. Exudatinite is a mobile or semi-mobile oil- or resin-like liptinite which has been recognized only in recent years (3). It occupies open spaces within the coal, and therefore tends to be found in fractures or

the lumens of coalified cells. Exudatinite in the Hanna 170 samples is especially
common in structured woody tissue (telinite) and fungal bodies (sclerotinite).
Inasmuch as exudatinite is rather mobile, and may actually migrate out of samples
into the immersion oil (4) Rich decided to emphasize exudatinite analyses. The
rationale was that, if any substances in the Hanna samples would react quickly to
thermal alteration, exudatinite would certainly be among them. Exudatinite is con-
fidently identified only in blue-light, as it fluoresces brightly, but is nearly
invisible in "white" light. In view of those facts, and because exudatinite is
believed to be of great potential value in geothermometry, Rich concentrated on
blue-light analyses.

Two pellets from each sample interval were counted to determine relative abundan-
ces of macerals. Each pellet was attached to a glass slide with modeling clay and
pressed onto the clay so that the observed surface lay parallel with the microscope
stage. Counts were performed along parallel transects which did not overlap.
Pellets were moved laterally by discrete units so that no adjacent fields of view
overlapped. One point was counted per field of view, that point lying at the inter-
section of an ocular cross-hair reticle. Five hundred points were counted per
pellet, and both pellets per sample were observed, providing 1000 points per sample.

TABLE 2

CHANGES IN LIPTINITE COMPOSITION FOR
SAMPLES FROM 277-278 FEET

Temperature°C	% Liptinites	# Exudatinite	# Liptodetrinite & Bituminite	# Exinite
19	4.4	10	17	10
50	4.9	12	22	8
100	3.6	14	14	4
150	2.8	8	8	5
200	3.7	8	22	5
250	4.0	5	17	5
300	2.8	1	13	6
350	1.0	1	7	0
400	0.0	0	0	0

TABLE 3

CHANGES IN LIPTINITE COMPOSITION FOR SAMPLES
FROM INTERVAL 284.2-285.2 FEET

Temperature°C	% Liptinites	# Exudatinite	# Liptodetrinite & Bituminite	# Exinite
21	7.7	1	58	10
50	8.5	3	67	9
100	11.5	4	87	12
150	9.3	7	68	14
200	8.3	4	69	5
250	8.5	2	67	7
300	5.2	1	38	8
350	0.8	0	6	2
400	0.0	0	0	0

RESULTS

Tables 2 and 3 illustrate several of the changes which took place during heating of samples from the 277-278 and 284.2-285.2 foot levels respectively. The following observations may be made:

1) In samples from both levels there was a gradual decrease in percentages of total liptinites as temperatures increased.

2) At both sample intervals, fluorescence essentially ceased at 400°C.

3) Reaction vesicles developed within vitrinite particles between 250-300°C. Vitrinite which contained spores or pollen, resin bodies, etc. became especially vesicular. Exudatinite showed distinct reactivity with consequent evacuation from sclerotinite bodies and interstices among crystals in pyrite framboids. Cracks developed in vitrinite.

4) Between 300-350°C vitrinite became obviously cracked and vesicular. Resinites clearly showed alteration rims as they reacted.

5) Between 350-400°C vitrinite became increasingly vesicular, with vesicle size and abundance increasing. Glassy or pitchey appearing deposits occurred in many vesicles. Exudatinite and resinite nearly vanished.

6) Within the 400-500°C range reflectivity of vitrinite particles appeared to be much more uniform. Resinite, exinite, cutinite, etc. were no longer visible in either "white" or blue light.

7) At 500-600°C, every coal particle developed a multitude of vesicles, and many were intensely cracked, almost brecciated. Coal particles were of uniform color and composition except for the vesicles and cracks.

Samples from the 279.7-280.7 foot interval were not point counted in blue light. The abundant clay particles were filled with detrital liptinites and were saturated with bituminite. Bituminite is another semi-mobile maceral, similar in some respects to exudatinite, which frequently infiltrates the layers of clay minerals. It thus causes the clays and interstices among clay particles to fluoresce. It is almost impossible to get an accurate quantification of bituminte in such clayey samples as virtually everything fluoresces, yet clearly the samples are not pure bituminite. Alteration in shale samples proceeded as listed for the other samples, however.

DISCUSSION AND CONCLUSIONS

Distinct petrographic changes did take place in the Hanna No. 1 samples as they were heated. Decreasing abundances of fluorescence macerals, increasing vesicularity and cracking in vitrinites, and eventual elimination of all macerals except vitrinite can be correlated with temperature ranges. It should, then, be feasible to take samples of carbonaceous shale and coal from UCG burn sites and determine the thermal gradiant away from the sites.

This work has also shown that macerals in the Hanna No. 1 coal react at quite different rates when heated. Liptinites are altered before vitrinites and intertinites, though eventually all the macerals become involved in the physical alterations. The fact that liptinites react earlier than vitrinites may carry important implications as far as maintaining optimum gas composition and production are concerned.

Liptinites, for example, are enriched in hydrogen as compared to the other maceral groups.   As liptinites decompose thermally, then, one would expect gases with a high hydrogen/carbon ratio to be produced. In a coal, such as the Hanna No. 1 which contains an abundance of easily altered liptinites (e.g. the resinite-rich Blind Canyon seam of Utah) conversion of coal to gas could occur at a comparatively low temperature and yet yield a gaseous product enriched in hydrogen. On the other hand, coals which do not have such an abundance of liptinites might require higher conversion temperatures which would involve more vitrinite in the conversion reactions in order to produce gases with a high hydrogen content.

An additional observation is that at higher temperatures, i.e., 400°C and above where particles become vesicular and fractured the reactive surface area within coal fragments is much greater than at cooler temperatures. The abundance of vesicles and the eventual brecciation of the particles should allow the coal to react more readily with hot gases within the gasification chamber. Brecciated coal particles would, of course, also allow product gases to move more readily through the coal bed than if the coal fragments were less extensively broken.

We believe the results of the work show that the most successful UCG operations will be those where careful consideration has been given to the petrographic com- position of the coals. Petrography can be used both as a geothermometer and as a technique to predict coal reactivity during underground gasification.

<div align="center">References</div>

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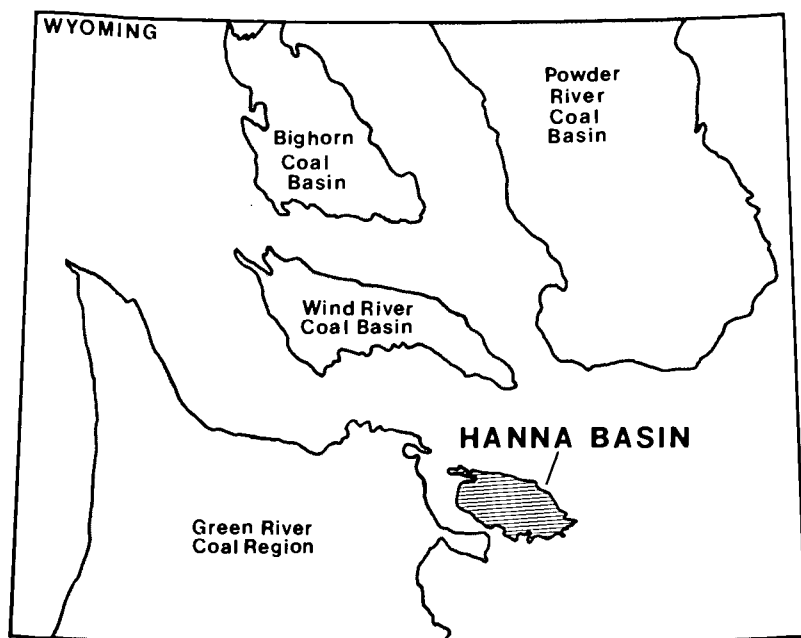
Figure 1
Location of Hanna Basin

WYOMING

Powder
River
Coal
Basin

Bighorn
Coal
Basin

Wind River
Coal Basin

HANNA BASIN

Green River
Coal Region

Figure 2
Bomb Assembly

thermocouple

steel-mesh hose

heat
tape

gas
path

crucible

furnace bomb

air trap bomb